TRIPLET-TRIPLET ENERGY TRANSFER IN DNA: A PROCESS THAT OCCURS ON THE NANOSECOND TIMESCALE

C. Curutchet,[1] A. A. Voityuk [1,2]

[1]Institut de Química Computacional, Universitat de Girona, Campus Montilivi, Girona, 17071, SPAIN

[2]Institució Catalana de Recerca i Estudis Avancats, Passeig Lluís Companys 23, Barcelona, 08010, SPAIN

In the past decade we have witnessed a notable progress in the comprehension of excite state dynamics and decay mechanisms in DNA.[1] Ultrafast laser spectroscopy experiments, complemented with powerful theoretical studies on the coupled nuclear and electronic degrees of freedom that dictate the dynamics in the system, have unveiled a detailed microscopic picture of the decay mechanisms in monomeric nucleobases. However, the corresponding dynamics in polynucleotides are still poorly understood, as assignment of observed decay rates are hampered by the uncertainty regarding energy transfer and delocalization of the corresponding excited states. While there have been several studies that address singlet excitation energy transfer through DNA stacks, mobility and delocalization of triplet excitons in DNA has yet to be ascertained.

In this contribution, triplet exciton migration in polyA–polyT DNA sequences is studied by using quantum-chemical methods coupled to classical molecular-dynamics simulations.[2] Environment effects on triplet-triplet electronic interactions are also examined based on the polarizable continuum model.[3] Our results indicate that triplet excited states in DNA are almost completely localized on single nucleobases, and their migration along the base stack occurs on the nanosecond timescale.

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[2] C. Curutchet, A. A. Voityuk, Angew. Chem. Int. Ed. 2011, 50, 1820-1822.
[3] C. Curutchet, A. A. Voityuk, Chem. Phys. Lett., submitted.